

What is claimed:

1. An indelible ink solution comprising:  
an organic solvent system;  
a dye or pigment dissolved in the organic solvent system; and  
an amino silane coupling agent.
2. The indelible ink of claim 1, wherein the amino silane coupling agent has the formula  
$$\text{Si}-(\text{R}_1)_4$$

Wherein

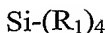
Each  $\text{R}_1$  is independently  $\text{R}_2$ ,  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ ,  $\text{C}_1-\text{C}_6\text{alkyl}$ , or Halogen, provided that at least one  $\text{R}_1$  is  $\text{R}_2$ ; and

Each  $\text{R}_2$  is independently  $\{\text{H}-(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})\}_m[(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})]_n$ -, in which  $m$  is 0 or 1 and  $n$  is 0 or 1.

3. The indelible ink of claim 2, wherein the amino silane coupling agent is  
 $\text{H}-(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})\}_m[(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})]_n-\text{Si}-(\text{R}_1)_3$ .
4. The indelible ink of claim 3, wherein at least two of  $\text{R}_1$  are halogen or  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ .
5. The indelible ink of claim 2, wherein the amino silane coupling agent is selected from  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane,  $\text{di}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylC}_1-\text{C}_6\text{alkylmonoamino}$  silane, and  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkyldiamino}$  silane.
6. The indelible ink of claim 1, wherein the amino silane coupling agent is a  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane.
7. The indelible ink of claim 1, wherein the amino silane coupling agent is  $\text{N}-\beta$  (aminoethyl)- $r$ -aminopropyl-trimethoxysilane,  $\text{N}-\beta$  (aminoethyl)- $r$ -aminopropyl-methyldimethoxysilane, 3-aminopropyl-triethoxysilane,  $\text{N}$ -phenyl- $r$ -aminopropyl-trimethoxysilane,  $\text{N}$ -( $n$ -Butyl)-3-aminopropyltrimethoxysilane, 3-aminopropylmethyldiethoxysilane.

8. The indelible ink solution of claim 1, wherein the solution includes between about 5 to about 30 weight percent of one or more amino silane coupling agents.
9. The indelible ink solution of claim 8, wherein the solution includes between about 15 to about 20 weight percent of one or more amino silane coupling agents.
10. The indelible ink solution of claim 1, further comprising a rheological modifier.
11. The indelible ink solution of claim 10, wherein the ink solution contains between about 0.1 and about 5 weight percent of the rheological modifier.
12. The indelible ink solution of claim 10, wherein the rheological modifier is a fumed silica.
13. The indelible ink solution of claim 1, wherein the dye or pigment is selected from Sepisol Fast Blue 2BR (*Solvent Blue 43*), Sepisol Fast Blue MBSN (*Solvent Blue 38*), Methyl Violet Base BP (*Solvent Violet 8 basic*), Sepisol Fast Blue ARNF (*Solvent Blue 37*), Sepisol Fast Blue 85219 (*Basic Blue 7 derivative*), Sepisol Fast Violet 881239 (*Basic Violet 1 derivative*), Sepisol Fast Violet 85152 (*Basic Violet 3 derivative*), Acid Blue 25, Acid Blue 158, and Basic Blue 54.
14. The indelible ink solution of claim 1, wherein the solution includes between about 15 to about 40 weight percent of one or more dyes in the aggregate.
15. The indelible ink solution of claim 1, wherein the ink solution when applied to paper remains visible when washed with one or more organic solvents.
16. A pen reservoir comprising:
  - a housing including an indelible ink solution including
  - an organic solvent system,
  - a dye or pigment dissolved or dispersed in the organic solvent system, and
  - an amino silane coupling agent.

17. The pen reservoir of claim 16, wherein the amino silane coupling agent has the formula



Wherein

Each  $\text{R}_1$  is independently  $\text{R}_2$ ,  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ ,  $\text{C}_1-\text{C}_6\text{alkyl}$ , or Halogen, provided that at least one  $\text{R}_1$  is  $\text{R}_2$ ; and

Each  $\text{R}_2$  is independently  $\{\text{H}-(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})\}_m[(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})]_n$ -, in which  $m$  is 0 or 1 and  $n$  is 0 or 1.

18. The pen reservoir of claim 17, wherein the amino silane coupling agent is  $\text{H}-(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})\}_m[(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})]_n\text{-Si}-(\text{R}_1)_3$ .

19. The pen reservoir of claim 18, wherein at least two of  $\text{R}_1$  are halogen or  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ .

20. The pen reservoir of claim 17, wherein the amino silane coupling agent is selected from  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane,  $\text{di}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylC}_1-\text{C}_6\text{alkylmonoamino}$  silane, and  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkyldiamino}$  silane.

21. The pen reservoir of claim 17, wherein the amino silane coupling agent is a  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane.

22. The pen reservoir of claim 17, wherein the amino silane coupling agent is  $\text{N}-\beta$  (aminoethyl)-*r*-aminopropyl-trimethoxysilane,  $\text{N}-\beta$  (aminoethyl)-*r*-aminopropyl-methyldimethoxysilane, 3-aminopropyl-triethoxysilane,  $\text{N}$ -phenyl-*r*-aminopropyl-trimethoxysilane,  $\text{N}$ -(*n*-Butyl)-3-aminopropyltrimethoxysilane, 3-aminopropylmethyldiethoxysilane.

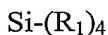
23. The pen reservoir of claim 16, wherein the solution includes between about 5 to about 30 weight percent of one or more amino silane coupling agents.

24. The pen reservoir of claim 23, wherein the solution includes between about 15 to about 20 weight percent of one or more amino silane coupling agents.

25. The pen reservoir of claim 16, wherein the ink solution further comprises a rheological modifier.
26. The pen reservoir of claim 25, wherein the ink solution contains between about 0.1 and about 5 weight percent of the rheological modifier.
27. The pen reservoir of claim 25, wherein the rheological modifier is a fumed silica.
28. The pen reservoir of claim 16, wherein the dye or pigment is selected from Sepisol Fast Blue 2BR (*Solvent Blue 43*), Sepisol Fast Blue MBSN (*Solvent Blue 38*), Methyl Violet Base BP (*Solvent Violet 8 basic*), Sepisol Fast Blue ARNF (*Solvent Blue 37*), Sepisol Fast Blue 85219 (*Basic Blue 7 derivative*), Sepisol Fast Violet 881239 (*Basic Violet 1 derivative*), Sepisol Fast Violet 85152 (*Basic Violet 3 derivative*), Acid Blue 25, Acid Blue 158, and Basic Blue 54.
29. The pen reservoir of claim 16, wherein the solution includes between about 15 to about 40 weight percent of one or more dyes in the aggregate.
30. The pen reservoir of claim 16, wherein the housing is capped at atmospheric pressure.
31. The pen reservoir of claim 16, wherein the housing is capped and pressurized to between about 3 to about 40 psig.
32. The pen reservoir of claim 31, wherein the housing is pressurized with a gas comprising at least 70% nitrogen.
33. The pen reservoir of claim 16, wherein the housing includes a ball point tip.
34. The pen reservoir of claim 16, wherein the ink solution when applied to paper remains visible when washed with one or more organic solvents.
35. A ball point pen comprising:  
a housing containing a ball point pen cartridge, wherein the ball point pen cartridge includes a ball point tip and an indelible ink solution, and wherein the indelible ink solution

includes an organic solvent system, a dye or pigment dissolved or dispersed in the organic solvent system; and an amino silane coupling agent.

36. The ball point pen of claim 35, wherein the amino silane coupling agent has the formula



Wherein

Each  $\text{R}_1$  is independently  $\text{R}_2$ ,  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ ,  $\text{C}_1-\text{C}_6\text{alkyl}$ , or Halogen, provided that at least one  $\text{R}_1$  is  $\text{R}_2$ ; and

Each  $\text{R}_2$  is independently  $\{\text{H}-(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})\}_m[(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})]_n$ -, in which  $m$  is 0 or 1 and  $n$  is 0 or 1.

37. The ball point pen of claim 36, wherein the amino silane coupling agent is  $\text{H}-(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})\}_m[(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})]_n\text{Si}-(\text{R}_1)_3$ .

38. The ball point pen of claim 37, wherein at least two of  $\text{R}_1$  are halogen or  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ .

39. The ball point pen of claim 36, wherein the amino silane coupling agent is selected from  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane,  $\text{di}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylC}_1-\text{C}_6\text{alkylmonoamino}$  silane, and  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkyldiamino}$  silane.

40. The ball point pen of claim 36, wherein the amino silane coupling agent is a  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane.

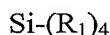
41. The ball point pen of claim 35, wherein the amino silane coupling agent is  $\text{N}-\beta$ -(aminoethyl)-*r*-aminopropyl-trimethoxysilane,  $\text{N}-\beta$ -(aminoethyl)-*r*-aminopropyl-methyldimethoxysilane, 3-aminopropyl-triethoxysilane,  $\text{N}$ -phenyl-*r*-aminopropyl-trimethoxysilane,  $\text{N}$ -(*n*-Butyl)-3-aminopropyltrimethoxysilane, 3-aminopropylmethyldiethoxysilane.

42. The ball point pen of claim 35, wherein the solution includes between about 5 to about 30 weight percent of one or more amino silane coupling agents.

43. The ball point pen of claim 42, wherein the solution includes between about 15 to about 20 weight percent of one or more amino silane coupling agents.
44. The ball point pen of claim 35, wherein the ink solution further comprises a rheological modifier.
45. The ball point pen of claim 44, wherein the ink solution contains between about 0.1 and about 5 weight percent of the rheological modifier.
46. The ball point pen of claim 44, wherein the rheological modifier is a fumed silica.
47. The ball point pen of claim 35, wherein the dye or pigment is selected from Sepisol Fast Blue 2BR (*Solvent Blue 43*), Sepisol Fast Blue MBSN (*Solvent Blue 38*), Methyl Violet Base BP (*Solvent Violet 8 basic*), Sepisol Fast Blue ARNF (*Solvent Blue 37*), Sepisol Fast Blue 85219 (*Basic Blue 7 derivative*), Sepisol Fast Violet 881239 (*Basic Violet 1 derivative*), Sepisol Fast Violet 85152 (*Basic Violet 3 derivative*), Acid Blue 25, Acid Blue 158, and Basic Blue 54.
48. The ball point pen of claim 35, wherein the solution includes between about 15 to about 40 weight percent of one or more dyes in the aggregate.
49. The ball point pen of claim 35, wherein the cartridge is capped at atmospheric pressure.
50. The ball point pen of claim 35, wherein the cartridge is capped and pressurized to between about 3 to about 40 psig.
51. The ball point pen of claim 35, wherein the cartridge is pressurized with a gas containing at least 70% nitrogen.
52. The ball point pen of claim 35, wherein the ink solution when applied to paper remains visible after washing with one or more organic solvents.
53. A method of recording a security marking comprising:

providing a writing instrument containing an indelible ink solution, wherein the indelible ink solution includes an organic solvent system, a dye or pigment dissolved in the organic solvent system; and an amino silane coupling agent.

54. The method of recording a security marking of claim 53, wherein the amino silane coupling agent has the formula



Wherein

Each  $\text{R}_1$  is independently  $\text{R}_2$ ,  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ ,  $\text{C}_1-\text{C}_6\text{alkyl}$ , or Halogen, provided that at least one  $\text{R}_1$  is  $\text{R}_2$ ; and

Each  $\text{R}_2$  is independently  $\{\text{H}-(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})\}_m[(\text{NH})-(\text{C}_1-\text{C}_6\text{alkyl})]_n\}$ -, in which  $m$  is 0 or 1 and  $n$  is 0 or 1.

55. The method of recording a security marking of claim 54, wherein the amino silane coupling agent is



56. The method of recording a security marking of claim 55, wherein at least two of  $\text{R}_1$  are halogen or  $\text{O}-(\text{C}_1-\text{C}_6\text{alkyl})$ .

57. The method of recording a security marking of claim 54, wherein the amino silane coupling agent is selected from  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane,  $\text{di}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylC}_1-\text{C}_6\text{alkylmonoamino}$  silane, and  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkyldiamino}$  silane.

58. The method of recording a security marking of claim 53, wherein the amino silane coupling agent is a  $\text{tri}(\text{C}_1-\text{C}_6\text{alkoxy})\text{C}_1-\text{C}_6\text{alkylmonoamino}$  silane.

59. The method of recording a security marking of claim 53, wherein the amino silane coupling agent is  $\text{N}-\beta$  (aminoethyl)-*r*-aminopropyl-trimethoxysilane,  $\text{N}-\beta$  (aminoethyl)-*r*-aminopropyl- methyltrimethoxysilane, 3-aminopropyl-triethoxysilane,  $\text{N}$ -phenyl-*r*-aminopropyl-trimethoxysilane,  $\text{N}$ -(*n*-Butyl)-3-aminopropyltrimethoxysilane, 3-aminopropylmethyltriethoxysilane.

60. The method of recording a security marking of claim 53, wherein the solution includes between about 5 to about 30 weight percent of one or more amino silane coupling agents.

61. The method of recording a security marking of claim 60, wherein the solution includes between about 15 to about 20 weight percent of one or more amino silane coupling agents.

62. The method of recording a security marking of claim 53, wherein the ink solution further comprises a rheological modifier.

63. The method of recording a security marking of claim 62, wherein the ink solution contains between about 0.1 and about 5 weight percent of the rheological modifier.

64. The method of recording a security marking of claim 62, wherein the rheological modifier is a fumed silica.

65. The method of recording a security marking of claim 53, wherein the dye or pigment is selected from Sepisol Fast Blue 2BR (*Solvent Blue 43*), Sepisol Fast Blue MBSN (*Solvent Blue 38*), Methyl Violet Base BP (*Solvent Violet 8 basic*), Sepisol Fast Blue ARNF (*Solvent Blue 37*), Sepisol Fast Blue 85219 (*Basic Blue 7 derivative*), Sepisol Fast Violet 881239 (*Basic Violet 1 derivative*), Sepisol Fast Violet 85152 (*Basic Violet 3 derivative*), Acid Blue 25, Acid Blue 158, and Basic Blue 54.

66. The method of recording a security marking of claim 53, wherein the solution includes between about 15 to about 40 weight percent of one or more dyes in the aggregate.

67. The method of recording a security marking of claim 53, wherein the writing instrument includes a cartridge that is capped at atmospheric pressure.

68. The method of recording a security marking of claim 53, wherein the cartridge is capped and pressurized to between about 3 to about 40 psig.



69. The method of recording a security marking of claim 53, wherein the cartridge is pressurized with a gas containing at least 70% nitrogen.

70. The method of recording a security marking of claim 53, wherein the ink solution when applied to paper remains visible when washed with one or more organic solvents.